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TAGUCHI ANALYSIS ON THE EFFECT OF PROCESS PARAMATERS ON DENSIFICATION DURING SPARK PLASMA SINTERING OF HfB2-20SiC (PREPRINT)

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14. ABSTRACT
Field assisted sintering (FAST) has emerged as a useful technique to densify ultra high temperature ceramics like HfB2-20SiC to a high density at relatively low temperatures and shorter times. The effect of various process variables on the densification during spark plasma sintering of HfB2-20SiC was studied using Taguchi analysis. The statistical analysis identified sintering temperature as the most significant parameter affecting the densification of HfB2-20SiC material. A density of 99% was achieved on sintering at 2100°C for 8 minutes at 30 kN pressure and heating rate of 100 K/min.

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Taguchi Analysis On The Effect Of Process Parameters On Densification During Spark Plasma Sintering Of HfB_2-20SiC

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Abstract

Field assisted sintering (FAST) has emerged as a useful technique to densify ultra high temperature ceramics like HfB_2-20SiC to a high density at relatively low temperatures and shorter times. The effect of various process variables on the densification during spark plasma sintering of HfB_2-20SiC was studied using Taguchi analysis. The statistical analysis identified sintering temperature as the most significant parameter affecting the densification of HfB_2-20SiC material. A density of 99% was achieved on sintering at 2100°C for 8 minutes at 30 kN pressure and heating rate of 100 K/min.

Introduction

Boride, carbides and nitrides of the group IVB and VB transition metals are considered ultrahigh temperature ceramics (UHTCs) based on melting temperature (typically in excess of 3000^{o}C) and other properties. Among the UHTCs, HfB_2 has a combination of properties such as chemical stability, high electrical and thermal conductivities, and resistance to erosion and corrosion. Subsequently, it has emerged as a potential candidate system for applications involving extreme chemical and high temperature thermal environments associated with hypersonic flight, atmospheric re-entry, and rocket propulsion [1-3]. However, HfB_2 exhibits poor oxidation resistance at temperatures above 1200°C. The oxidation resistance of HfB_2 is increased with addition of 20-30 vol. % SiC [3-5].

UHTCs typically require external pressure, high temperature and long sintering cycles for densification due to the properties of strong covalent bonds and low self-diffusion [6]. In recent years, various studies were carried out utilizing the spark plasma sintering (SPS) technique for densifying the UHTCs at relatively lower temperature and shorter times [7-12]. In SPS technique the material is densified under the influence of external uniaxial pressure and pulsed direct current.

Few studies have been reported regarding SPS sintering of HfB_2- SiC composites [1, 2, 6-9]. However these studies were primarily focused on investigating the ability of SPS technique to achieve full density HfB_2- SiC samples. There was no study carried out to understand the effect of various processing parameters on densification of HfB_2- SiC during SPS sintering. The goal of the present research is to understand and identify critical process variables effecting the densification of HfB_2- 20SiC during SPS.

Experimental

Ball milled HfB_2 (-325 mesh Cerac, USA) and SiC powders (1 μm, H.C. Starck, USA) were used as the starting powders. The powders were loaded into a 40-mm graphite die coated with BN and lined with graphite foil. The samples were sintered using FAST (FCT Systeme GmbH, Rauenstein, Germany). The temperature was measured by an optical pyrometer focused on the bottom of a bore hole in the graphite punch approx. 5 mm from the powder. A vacuum of 150 Pa was maintained for the entire heating cycle. A pulsed DC current of 15 ms on and 5 ms off with a single pulse was used for heating. Experiments were designed based on Taguchi analysis concepts was carried out to understand the effect of various processing parameters of spark plasma sintering on the densification of HfB_2-20SiC. The effect of processing parameters including temperature, hold time, holds pressure and heating rate was studied. The L_9 orthogonal array and the levels of the variables investigated in the current study are summarized in **Table 1**. The data analysis of the results was carried out using MINITAB software.

Results and Discussion

The results obtained from the SPS sintering experiments are summarized in **Table 2**. The density of the samples after sintering was monitored to quantify the effect of various process parameters. The output characteristic (sintered density) can be analyzed in three categories i.e. the lower-the better, the nominal-the- better or the higher-the-better [15,16].

In the current analysis a high sintered density is desirable thus the statistical analysis is carried out with higher-the-better option. Taguchi analysis utilizes signal-to-noise (S/N) ratio to quantify the quality characteristics deviating from the desired values. The larger S/N ratio corresponds to better quality characteristics. The S/N ratio for the higher-the-better type is estimated as per **Equation 1**

$$\left(\frac{S}{N}\right)_{HB} = -10\log\left[\frac{1}{R}\sum_{J=1}^{R}\frac{1}{Y_j^2}\right] \qquad (1)$$

Where Yj, $j = 1, 2 \ldots n$ are the response values and R is the number of repetitions. The S/N values of the SPS sintering runs are shown in **Table 2**.

The main effects plot showing the variation in sintered density with processing parameters is shown in **Figure 1.** The main effects plot showing the variation in S/N ratio with processing parameters is shown in **Figure 2**. The main effects plots show an increase in density of samples with temperature. The highest density was obtained at a sintering temperature of 2373K (2100^{o}C). The density of the sample also was found to increase with increase in hold time from 2 to 8 minutes. However the increase in density with further increase in time form 8 to 14 minutes was marginal. The heating rate showed an inverse effect on the sintered density. Increase in heating rate resulted in lower sintered density. A similar effect of heating rate on density was reported during SPS sintering of Al_2O_3 [17, 18] and $BaTiO_3$ [19]. The high density at low heating rates is attributed to small grain size of the samples. The small grain size provides large grain boundary area enhancing the grain boundary diffusion phenomena resulting in higher densification of the sample. This phenomenon may become more effective in the case of SPS sintering process where densification occurs due to the high temperature sintering mechanisms of grain boundary and volume diffusions. The SEM images for sintered samples from Run 8 and Run 9 are shown in **Figure 3** and **Figure 4** respectively. The SEM micrographs of samples sintered at various conditions showed SiC (dark) and HfB_2 (grey) phases.

Analysis of variance (ANOVA) was carried out to estimate the significance and contribution of each parameter to the overall sinter density. The ANOVA analysis on the sintered density is shown in **Table 3**. The analysis in **Table 3** identifies temperature as the primary parameter having significant effect on the sintered density. The contribution of temperature to sintered density was 64.85%. In contrast to this result, the Taguchi analysis of plasma pressure compaction of a nanoscale SiC system revealed a more equitable dependence on the four process parameters [20]. After temperature the time and heating rate were major parameters affecting the sintered density. The contribution of time and heating rate to sinter density was 21.29% and 13.37% respectively. The data analysis from the study identified sintering temperature and time as critical parameters having a significant effect on densification of HfB_2-20SiC. The analysis can be further extended to identify the critical process parameters effecting the sinter densification of various UHTC materials.

Conclusions

The effect of various process variables sintering temperature, time, heating rate and hold on pressure on the densification during spark plasma sintering of HfB_2-20SiC was studied using Taguchi analysis. A density of 99% was achieved on sintering at 2100°C for 8 minutes at 30 kN pressure and heating rate of 100 K/min. The statistical analysis identified sintering temperature, time and heating rate as the major processing variables affecting the sinter density of the samples. The contribution of temperature and time on sintered density of the sample was estimated as 64.85 and 21.29% respectively.

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Process parameters	Range	Level 1	Level 2	Level 3
Temperature (K)	2173-2373	1900	2000	2100
Hold time (min)	2-14	2	8	14
Hold pressure (kN)	30-50	30	40	50
Heating rate (K/min)	50-150	50	100	150

Table 1. Process parameters and selected levels in the current study.

Run	Temperature (K)	Hold time, (min)	Hold pressure, (kN)	Heating rate, (K/min)	Density, (g/cc)	Density*, % theoretical	S/N
1	2173	2	30	50	7.49	84.3	17.49
2	2173	8	50	150	7.44	83.8	17.43
3	2173	14	40	100	7.94	89.4	17.99
4	2273	2	40	100	7.54	84.9	17.54
5	2273	8	50	50	8.20	92.4	18.28
6	2273	14	30	150	7.83	88.2	17.88
7	2373	2	40	150	8.05	90.7	18.12
8	2373	8	30	100	8.79	99	18.88
9	2373	14	50	50	8.72	98.2	18.81

Table 2. Summary of the results obtained from the SPS sintering runs.

	Sum of Squares	Degrees of freedom	Significance
Temperature	1.31	2	64.85%
Time	0.43	2	21.29%
Pressure	0.01	2	0.50%
Rate	0.27	2	13.37%
Total	2.01	8	100%

Table 3. ANOVA results showing the significance and contribution of each processing parameter on the sintered density.

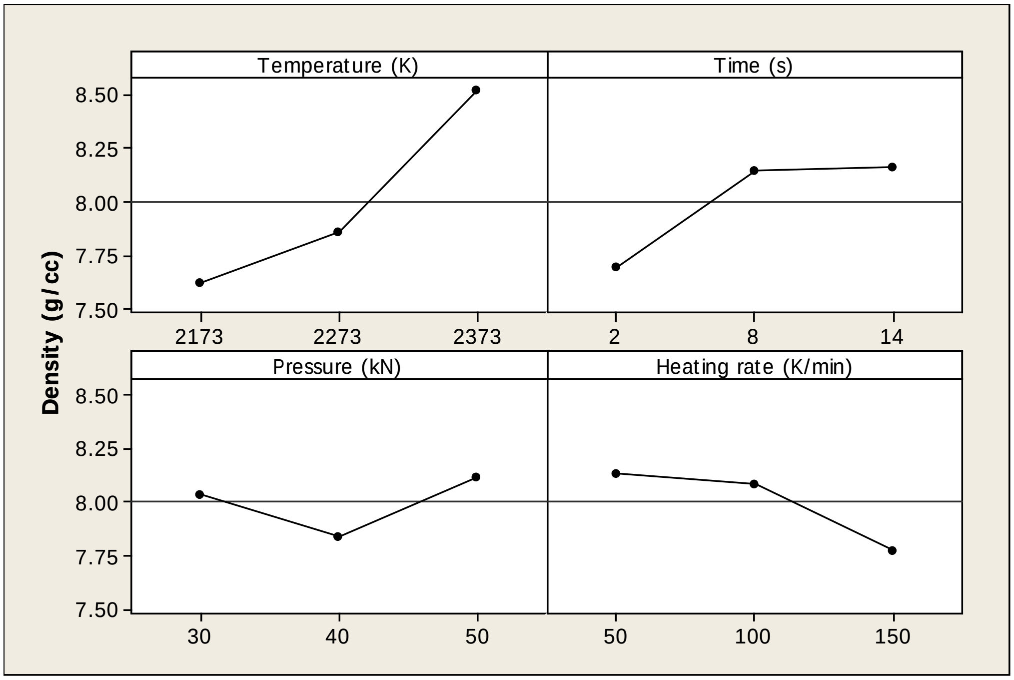

Figure 1: Main effects plot showing the effect of processing parameters on sintered density.

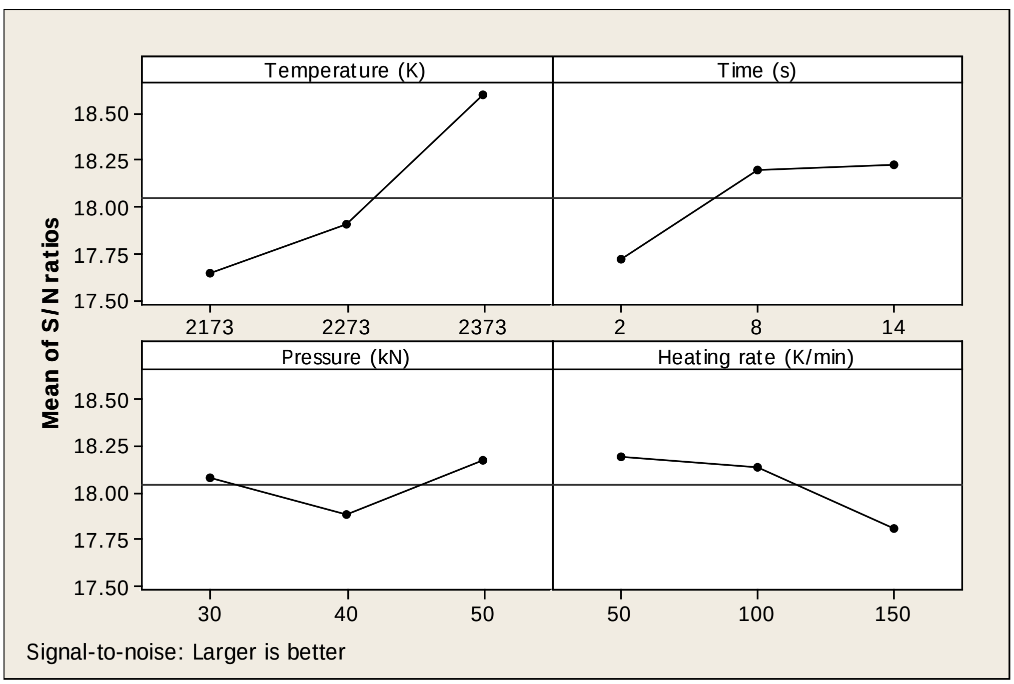

Figure 2: Main effects plot showing the effect of processing parameters on S/N ratio.

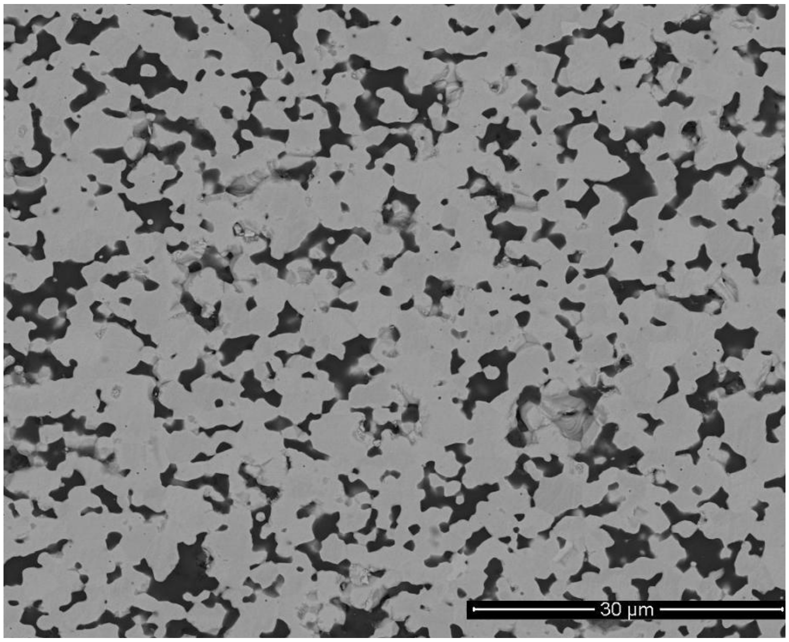

Figure 3: SEM and image analysis data of HfB_2-20SiC samples sintered as per Run 8 conditions (2373K, 14s, 50kN, 50K/min)

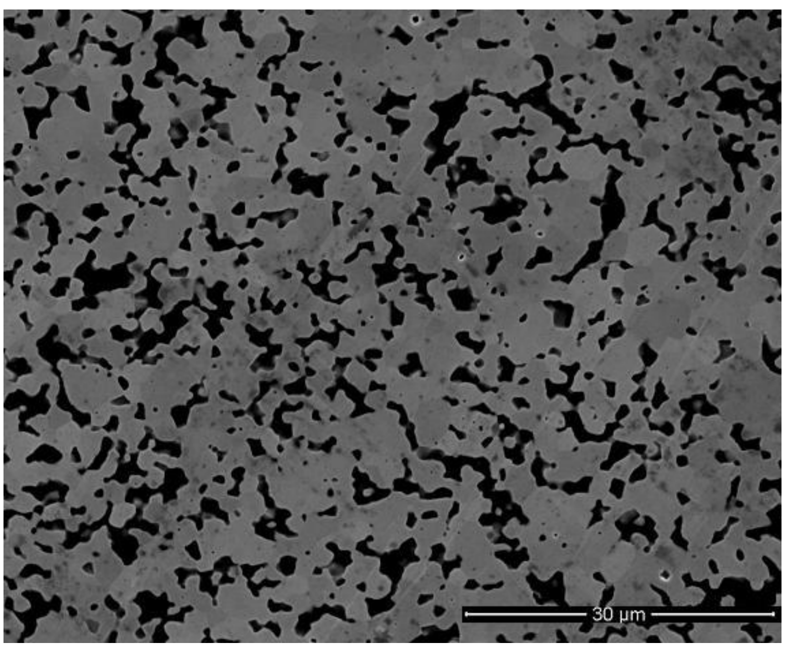

Figure 4: SEM and image analysis data of HfB_2-20SiC samples sintered as per Run 9 conditions (2373K, 8 s, 30kN, 100K/min)